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MOTOR VEHICLES AS SOURCES OF COMPOUNDS IMPORTANT TO TROPOSPHERIC AND STRATOSPHERIC OZONE

by

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16. ABSTRACT

One of the most rapidly growing human activities in the U.S. of importance to atmospheric ozone is the use of highway motor vehicles. Transportation sources are estimated to have been responsible for about 34% of 1985 U.S. anthropogenic hydrocarbon emissions, 70% of carbon monoxide emissions, 45% of nitrogen oxide emissions, 24% of nonaerosol chlorofluorocarbon emissions, and 14% of carbon dioxide emissions. Data is presented describing possible uninventoried transportation hydrocarbon emissions that could increase their estimated contribution to 45 - 50% of the anthropogenic total. Data is also presented suggesting motor vehicles to be relatively insignificant sources of anthropogenic nitrous oxide, but noting that these emissions are increased by the control technologies used to reduce hydrocarbon, carbon monoxide, and nitrogen oxides emissions. The sensitivity of motor vehicle emission rates and compositions to such operating variables as ambient temperature, altitude, and average speed is discussed. Hydrocarbon and carbon monoxide emission rates are generally minimized at ambient temperatures of about 24°C (75°F), and elevated at higher and lower temperatures. These emissions are also elevated by high altitude and reduced average speed operating conditions. Nitrogen oxide emission rates are not as sensitive to vehicle operating conditions. The ratio of hydrocarbon to nitrogen oxide emissions, of importance to tropospheric ozone chemistry, varies with operating conditions from about 1 to 5.

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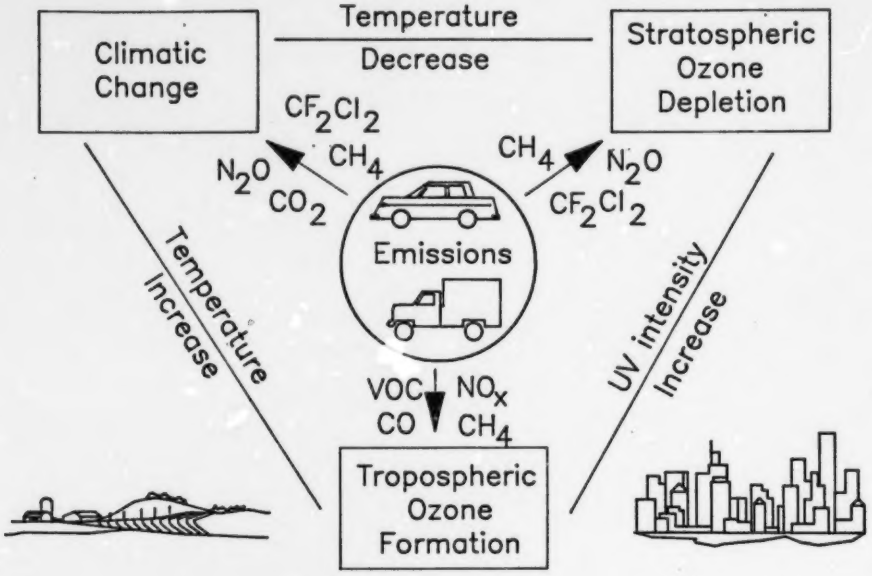
Motor Vehicles as Sources of Compounds -
Important to Tropospheric and Stratospheric Ozone



MOTOR VEHICLES AS SOURCES OF COMPOUNDS IMPORTANT TO TROPOSPHERIC AND STRATOSPHERIC OZONE

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ABSTRACT

One of the most rapidly growing human activities in the U.S. of importance to atmospheric ozone is the use of highway motor vehicles. Transportation sources are estimated to have been responsible for about 34% of 1985 U.S. anthropogenic hydrocarbon emissions, 70% of carbon monoxide emissions, 45% of nitrogen oxide emissions, 24% of nonaerosol chlorofluorocarbon emissions, and 14% of carbon dioxide emissions. Data is presented describing possible uninventoried transportation hydrocarbon emissions that could increase their estimated contribution to 45 - 50% of the anthropogenic total. Data is also presented suggesting motor vehicles to be relatively insignificant sources of anthropogenic nitrous oxide, but noting that these emissions are increased by the control technologies used to reduce hydrocarbon, carbon monoxide, and nitrogen oxides emissions. The sensitivity of motor vehicle emission rates and compositions to such operating variables as ambient temperature, altitude, and average speed is discussed. Hydrocarbon and carbon monoxide emission rates are generally minimized at ambient temperatures of about 24°C (75°F), and elevated at higher and lower temperatures. These emissions are also elevated by high altitude and reduced average speed operating conditions. Nitrogen oxide emission rates are not as sensitive to vehicle operating conditions. The ratio of hydrocarbon to nitrogen oxide emissions, of importance to tropospheric ozone chemistry, varies with operating conditions from about 1 to 5. The composition of hydrocarbon emissions varies with vehicle speed and ambient temperature; elevated paraffinic fractions are typical of high speed and high temperature operation, and elevated olefinic fractions are typical of low speed and low temperature operation.

INTRODUCTION

Since enactment of the 1970 Clean Air Act, ozone has proven to be the most pervasive and difficult to control of all U.S. air pollutants. Unlike the other pollutants addressed by this Act, ozone is not directly emitted from sources; rather, it results from complex photochemical reactions involving sunlight and various precursor emissions that emanate from both man's activities and natural processes. Atmospheric transport has complicated efforts to control ozone as authorities charged with maintaining local air quality are often frustrated by ozone precursors resulting from activities hundreds of miles upwind of their jurisdiction. The phenomenon of pollutant transport has also awakened a growing international concern about changes in stratospheric air chemistry wherein the rate of ozone destruction is being

accelerated by compounds transported from the earth's surface. Both tropospheric and stratospheric temperature change can also be associated with emissions at the earth's surface. The chemistry of the earth's atmosphere is very complex and the subject of much hypothesizing and study. It is apparent that the tropospheric and stratospheric systems are coupled and should be collectively considered when examining the impact of man's activities on the atmosphere.

One of the most rapidly growing anthropogenic activities of importance to atmospheric ozone is the use of highway motor vehicles. Lee Thomas, Administrator of the U.S. Environmental Protection Agency (USEPA), recently noted that since 1970 the U.S. economy grew about 44 percent, the population 18 percent, and the kilometers driven by American motorist 58 percent (ref. 1) There are many compounds emitted from motor vehicles that participate in atmospheric chemical and physical processes which influence the concentrations of tropospheric and stratospheric ozone. Volatile organic compounds (VOC) and nitrogen oxides (NOx or NO + NO_2) are the primary precursors of tropospheric ozone. Methane (CH_4) and carbon monoxide (CO) reactions control the atmospheric concentrations of hydroxyl radical (OH) which in turn controls the atmospheric residence time of compounds important to radiative heating of the troposphere (green house effect) and compounds important to ozone chemistry including stratospheric ozone depletion. CO, although of lower reactivity than most VOCs, is also considered a precursor of tropospheric ozone. The nitrous oxide (N_2O), Freon-12 (CF_2Cl_2), and CH_4 transported from the earth's surface to the stratosphere participate in ozone depletion chemistry. A product of depleted stratospheric ozone is more intense UV radiation in the troposphere where it drives the formation of ozone. Ozone, N_2O, CH_4, CF_2Cl_2, and carbon dioxide (CO_2) also all influence the radiative balance of the earth's atmosphere. Increased atmospheric concentrations of these compounds will elevate the temperature of the troposphere and decrease the temperature of the stratosphere. These temperature changes can influence the rates of reactions forming and destroying ozone.

MOTOR VEHICLE EMISSIONS

The atmospheric concentrations of CF_2Cl_2, N_2O, CO, CO_2, and CH_4 are currently increasing at annual rates of approximately 5%, 0.2%, 1-2%, 0.5%, and 1%, respectively (ref. 2). There are many anthropogenic and biogenic sources of most of these compounds. The relative strengths of the varied sources are often debated and are important to the potential impact of anthropogenic emissions control. Only highway motor vehicle sources will be discussed in this presentation. The U.S. government has taken measures to

reduce ambient levels of VOC, NOx, CO, and indirectly CO_2, through motor vehicle emissions (tailpipe and evaporative) and fuel economy regulations. Table 1 summarizes some of these regulations.

Countering the government mandated emission rate reductions have been the previously mentioned dramatic growth of vehicle-kilometers-traveled (VKT) and a disturbingly large amount of vehicle emission control system tampering. Fig. 1 illustrates U.S. light-duty and heavy-duty vehicle urban and rural VKT growth during the period 1975 to 1985, with projections through 2000 (ref. 3). During the 1975 to 1985 decade, light-duty urban and rural VKT increased 26.8% and 29.2%, respectively, and heavy-duty urban and rural VKT increased 53.0% and 61.1%, respectively. The results of the 1986 USEPA Motor Vehicle Tampering Survey (15 cities, 7541 vehicles) suggested that one out of every five passenger cars and light-duty trucks in the U.S. had at least one component of the emission control system tampered (ref. 4). An additional 25% were classified as "arguably tampered," meaning that a determination could not be made as to whether the vehicle's condition was due to tampering or poor maintenance. Tampering and misfueling can cause dramatic increases in VOC, CO, and NOx emissions. For example, disconnected air pumps (found on 8% of the surveyed vehicles so equipped) can increase VOC emissions 200% and CO emissions 800%, and disconnected exhaust-gas-recirculation systems (found on 7% of surveyed vehicles so equipped) can increase NO_x emissions 175%. Misfueling catalyst equipped vehicles with leaded gasoline (found on 9% of vehicles requiring unleaded fuel) can increase VOC emissions 500% and CO

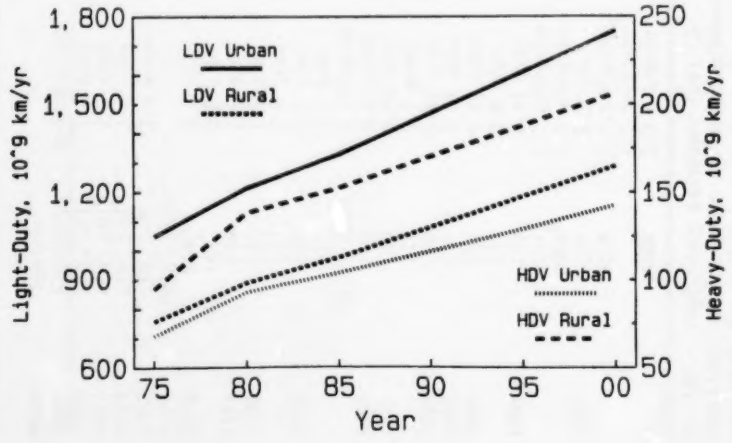


Fig. 1. U.S. motor vehicle kilometers traveled, 1975 to 2000.

TABLE 1
U.S. motor vehicle emissions and fuel economy regulations.

Model Year	Vehicle[1] Category	Hydrocarbons	Carbon Monoxide	Oxides of Nitrogen	Hydrocarbons + Oxides of Nitrogen	Evaporative Hydrocarbon	Fuel Economy
1970	LDV, LDT[2]	2.2 g/mi	23 g/m	-	-	-	-
1971	LDV, LDT[3]	2.2 g/mi	23 g/mi			6 g/test	-
1972	LDV, LDT[4]	3.4 g/mi	39 g/mi	-	-	2 g/test	-
1973	LDV, LDT[5]	3.4 g/mi	39 g/mi	3.0 g/mi	-	2 g/test	-
1974	HDGV, HDDV	-	40 g/bhp-hr	-	16 g/bhp-hr	-	-
1975	LDV[6]	1.5 g/mi	15 g/mi	3.1 g/mi	-	2 g/test	-
	LDT[6]	2.0 g/mi	20 g/mi	3.1 g/mi	-	2 g/test	-
1977	LDV	1.5 g/mi	15 g/mi	2.0 g/mi	-	2 g/test[7]	-
1978	LDV	1.5 g/mi	15 g/mi	2.0 g/mi	-	6 g/test[7]	18 mpg
	LDT	2.0 g/mi	20 g/mi	3.1 g/mi	-	6 g/test	-
1979	LDV	1.5 g/mi	15 g/mi	2.0 g/mi	-	6 g/test	19 mpg
	LDT	1.7 g/mi	18 g/mi	2.3 g/mi	-	6 g/test	-
	HDGV, HDDV	1.5 g/bhp-hr	25 g/bhp-hr	-	10 g/bhp-hr	-	-
1980	LDV	0.41 g/mi	7.0 g/mi	2.0 g/mi		6 g/test	20 mpg
1981	LDV	0.41 g/mi	3.4 g/mi	1.0 g/mi	-	2 g/test	22 mpg
	LDT	1.7 g/mi	18 g/mi	2.3 g/mi	-	2 g/test	-
1982	LDV	0.41 g/mi	3.4 g/mi	1.0 g/mi	-	2 g/test	24 mpg
1983	LDV	0.41 g/mi	3.4 g/mi	1.0 g/mi	-	2 g/test	26 mpg
1984	LDV	0.41 g/mi	3.4 g/mi	1.0 g/mi	-	2 g/test	27 mpg
	LDT	0.8 g/mi	10 g/mi	2.3 g/mi	-	2 g/test	-
1985	LDV[8]	0.41 g/mi	3.4 g/mi	1.0 g/mi	-	2 g/test	27.5 mpg
	HDGV[8]	2.5 g/bhp-hr	40 g/bhp-hr		10.7 g/bhp-hr	3 g/test, 4 g/test[9]	-
	HDDV[8]	1.3 g/bhp-hr	15.5 g/bhp-hr		10.7 g/bhp-hr		-
1987	LDT	0.8 g/mi	10 g/mi	1.2 g/mi	-	2 g/test	-
	HDGV	1.3 g/bhp-hr[9]	15.5 g/bhp-hr	6.0 g/bhp-hr	-	3 g/test	-
		2.5 g/bhp-hr	40 g/bhp-hr	6.0 g/bhp-hr		4 g/test	
	HDDV	1.3 g/bhp-hr	15.5 g/bhp-hr	6.0 g/bhp-hr			

1). LDV: light-duty passenger car; LDT: light-duty truck; HDGV: heavy-duty gasoline truck/bus; HDDV: heavy-duty diesel truck/bus, g/mi = grams per mile; g/bhp-hr = grams per brake horsepower hour, mpg = miles per gallon. 2). 7-mode test procedure for tailpipe emissions 3). carbon trap procedure for evaporative emissions 4). CVS-72 test procedure for tailpipe emissions 5). 13-mode test procedure for tailpipe emissions 6). CVS-75 test procedure for tailpipe emissions 7). SHED procedure for evaporative emissions 8). transient test procedure for tailpipe emissions 9). vehicles larger than 14,000 pounds gross-vehicle-weight

emissions 400%. The survey indicated that locally administered Inspection and Maintenance Programs and Antitampering Programs reduced tampering and misfueling by 35% and 50%, respectively.

Highway motor vehicles are but one category of anthropogenic source. The combined impact of motor vehicle emissions regulations, VKT growth, and tampering, and the relative importance of transportation and other anthropogenic sources are illustrated in Figs. 2,3, and 4 for VOC, CO, and NOx, respectively, for the period 1940 to 1985 (ref. 5). U.S. emissions peaked during the early 1970's. Since then, NOx emissions have remained relatively constant, and VOC and CO emissions have decreased. Transportation as an anthropogenic source of VOC has varied from 28.3% in 1940, to 45.6% in 1970, to 33.8% in 1985. Similar observations suggest that transportation accounted for 35.9% of CO in 1940, 72.7% in 1970, and 70.4% in 1985, and accounted for 32.4% of NOx in 1940, 42.0% in 1970, and 44.2% in 1985. In 1985, the estimated national transportation VOC, CO, and NOx mass emission rates were 7.2 million metric tons/yr, 47.5 million metric tons/yr, and 8.[illegible] million metric tons/yr, respectively. This data was taken from the National Air Quality and Emissions Trends Report, 1985, published February, 1987, which used the EPA computer model MOBILE 3 to estimate transportation contributions. The current developmental version of this model (to be released as MOBILE 4) will increase the estimated contribution of transportation sources to the nationwide VOC inventory. For example, using MOBILE 4, USEPA recently estimated that motor vehicles contributed 48% of the 1983 total U.S. non-

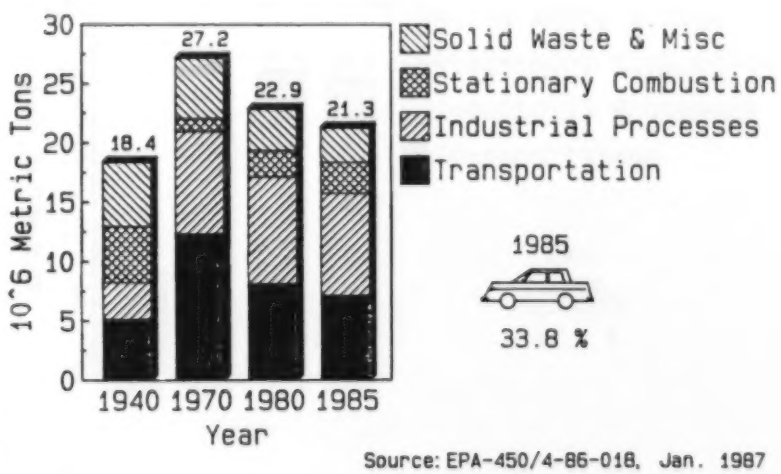
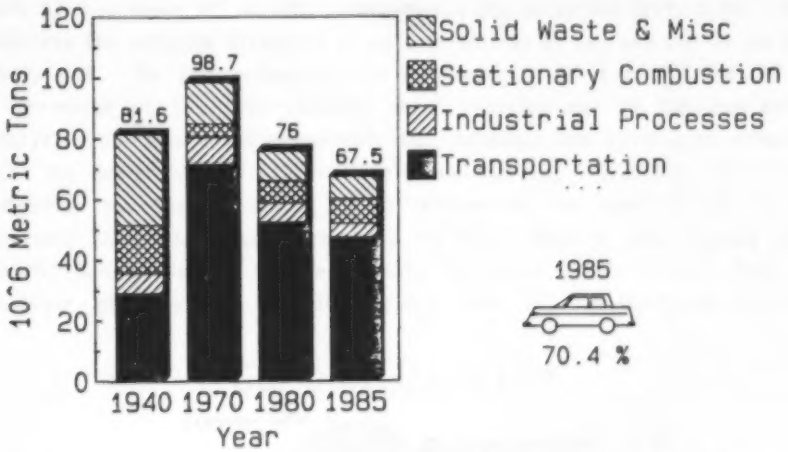


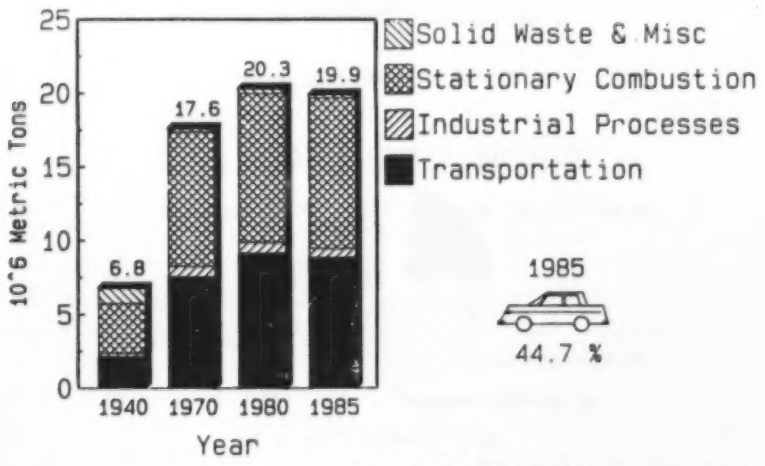
Fig. 2. Volatile organic compound emissions trend, 1940 to 1985.

methane hydrocarbon inventory (ref. 6). The differences between MOBILE 3 and MOBILE 4 VOC estimates will be discussed in detail later. The most significant change involves the use of area-specific gasoline volatilities and ambient temperatures which substantially increases the magnitude of evaporative emission estimates during the high temperature periods of the summer



Source: EPA-450/4-86-018, Jan. 1987

Fig. 3. Carbon monoxide emissions trend, 1940 to 1985.



Source: EPA-450/4-86-018, Jan. 1987

Fig. 4. Nitrogen oxide emissions trend, 1940 to 1985.

months. CO and NOx emission estimates were not significantly changed.

Data is not as readily available for unregulated emissions such as N_2O, CO_2, and CF_2Cl_2. However, estimates can be made of the relative strengths of the anthropogenic sources of these compounds. The transportation sector accounted for less than 10% of 1985 anthropogenic N_2O emissions (ref. 7), about 24% of 1985 CFC emissions, i.e. nonaerosol CFC-11, CFC-12, and CFC-113 (ref. 8), and about 14% of 1985 anthropogenic CO_2 emissions (ref. 9,10). Fig. 5 depicts the relative strengths of various sources of CO_2 and CFC in the U.S. during 1985. The 1985 transportation mass emission rates of N_2O and CFC can be estimated at about 80 thousand metric tons/yr and 48 thousand metric tons/yr, respectively. Anthropogenic CO_2 emissions are associated primarily with the combustion of fossil fuels and bio-mass. Globally, fossil fuel combustion has been estimated to be responsible for about 50-70% of CO_2 emissions (ref. 9). DeLuchi recently estimated that in 1985 highway motor vehicle fuel combustion was responsible for about 14% of fossil fuel CO_2 emissions globally and 24% in the U.S. (ref. 10). He estimated that U.S.

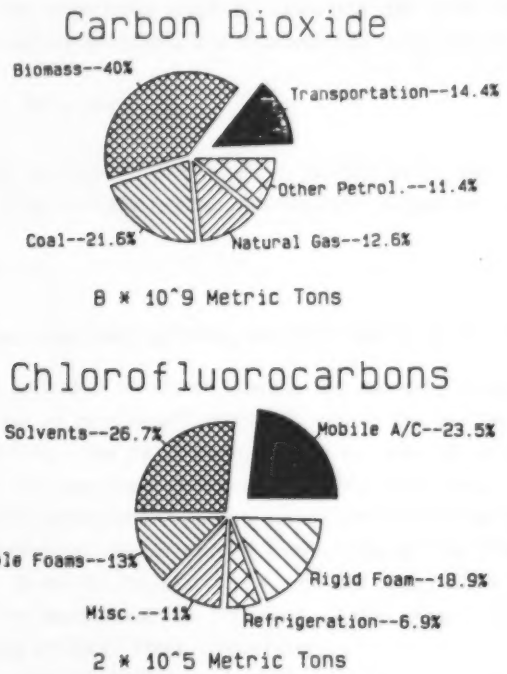


Fig. 5. 1985 carbon dioxide and nonaerosol chlorofluorocarbon emissions by source category.

highway motor vehicles emitted about 1.1 billion metric tons of CO_2 during 1985.

VOC, CO, NOx emissions

Motor vehicle emissions are sensitive to the type of engine, age, maintenance, size of the vehicle, type of fuel being used, and among other things, the conditions under which the vehicle is operated, e.g. speed, altitude, and ambient temperature. Depending on these considerations, the composition and rate of emissions will vary. The USEPA maintains a model (MOBILE 3 current release, MOBILE 4 in development) useful for predicting fleet average VOC, CO, and NOx emission characteristics as a function of many of these variables (ref. 11). Table 2 provides an example of the information that can be obtained from this model. Seven categories of motor vehicles are incorporated. For each calendar year and vehicle category, the VKT and emissions are distributed over 20 model years. The model incorporates tailpipe emissions, g/km equivalents of evaporative emissions, and g/km equivalents of refueling emissions (in the developmental version) in the THC emission rates. The algorithms used to calculate the g/km equivalents of evaporative and refueling emissions are based on the relationships:

$$\text{Evap., g/km} = (Di + TPD \times Hs) / KPD \quad (1)$$

where Di = diurnal emissions, g/day; TPD = average trips per day; Hs = hot soak emissions, g/trip; and KPD = average kilometers driven per day, and

$$\text{Refuel., g/km} = RF / FE \quad (2)$$

where RF = refueling emissions, g/liter; and FE = fuel economy, km/liter.

Mobile 3 used 3.05 and 50.0 for fleet average TPD and KPD, respectively. The developmental version of this model, MOBILE 4, varies TPD and KPD according to the age of the vehicle. The tailpipe rates can be reported as THC emissions (as regulated), or as nonmethane hydrocarbon (NMHC) emissions. As indicated in Table 2, emission characteristics vary among vehicle categories, and fleet average values, therefore, depend on the composition of the fleet, i.e. the VKT mix. The mix given in Table 2 represents the model default values for 1985 and are used for most scenarios discussed in this paper. This mix can be varied to correspond to local fleet compositions.

Fig. 6 illustrates how MOBILE 4 characterizes fleet average THC, CO, and NOx emissions for the period 1975 through 2000, at 152 m (500 ft) altitude, 24°C (75°F), 32 km/h (20 mi/h) average speed, with Inspection and Maintenance

TABLE 2
1985 motor vehicle fleet average emissions factors [1]

	LDGV[2]	LDGT	HDGV	LDDV	LDDT	HDDV	MC	ALL
Veh. Spd, km/h	32.2	32.2	32.2	32.2	32.2	32.2	32.2	
VKT Mix	0.652	0.218	0.040	0.023	0.008	0.054	0.007	
Exhaust NMHC, g/km	1.06	2.08	3.20	0.24	0.38	2.80	2.22	1.45
Exhaust HC, g/km	1.14	2.19	3.44	0.25	0.39	2.88	2.37	1.54
Evap. HC, g/km	0.86	1.33	4.51	-	-	-	2.11	1.05
Refuel. HC, g/km	0.21	0.27	0.44	-	-	-	-	0.21
THC, g/km	2.21	3.79	8.37	0.25	0.39	2.88	4.48	2.79
CO, g/km	11.95	20.96	86.74	0.80	0.96	7.76	12.40	16.33
NOx, g/km	1.24	2.09	3.46	0.86	1.02	12.84	0.52	2.13

[1] altitude 152 m, ambient temperature 23.9°C, with Inspection and Maintenance/Antitampering Programs

[2] LDGV: light-duty gasoline vehicle; LDGT: light-duty gasoline truck; HDGV: heavy-duty gasoline vehicle; LDDV: light-duty diesel vehicle; LDDT: light-duty diesel truck; HDDV: heavy-duty diesel vehicle; MC: motorcycle

(I&M) and Anti-Tampering Programs (ATP). From 1975 to 1985 fleet average THC emission rates were reduced 57%, CO emission rates 62%, and NOx emissions rates 35%. The indicated emission rates were determined using laboratory simulations of urban driving conditions as used in Federal Emissions

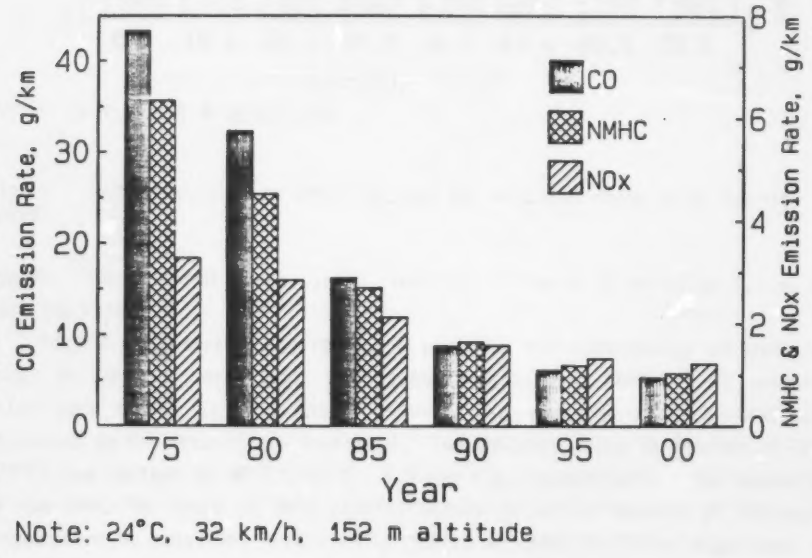
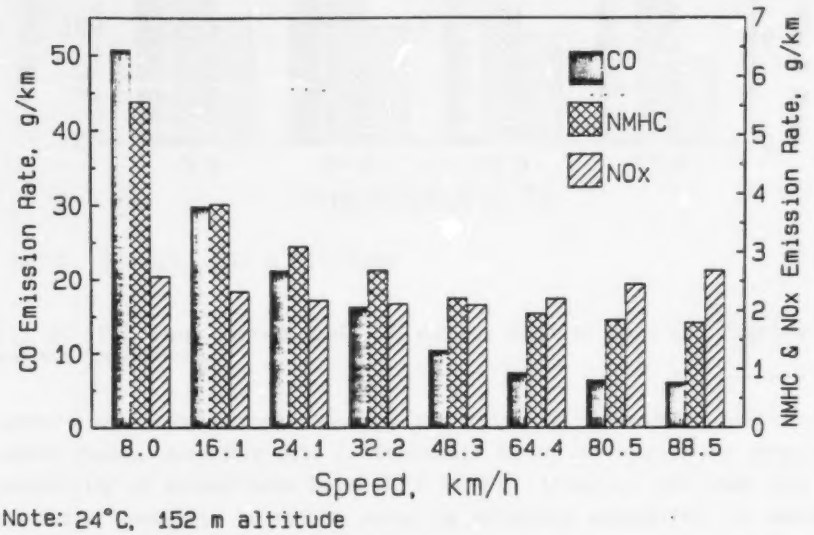


Fig. 6. Fleet average NMHC, CO, and NOx emission rates, 1975 to 2000.

Certification (ref. 12).

Fig. 7 illustrates how MOBILE 4 predicts fleet average emission rates as a function of vehicle speed. NMHC and CO emission rates continuously decrease as average speed is increased. The 1985 fleet average NMHC and CO rates at 88 km/h (55 mi/h) were 32% and 12%, respectively, of the values at 8-km/h (5 mi/h). The NOx fleet average emission rate passed through a minimum in the 32 - 48 km/h (20 - 30 mi/h) range, and was elevated at both lower and higher

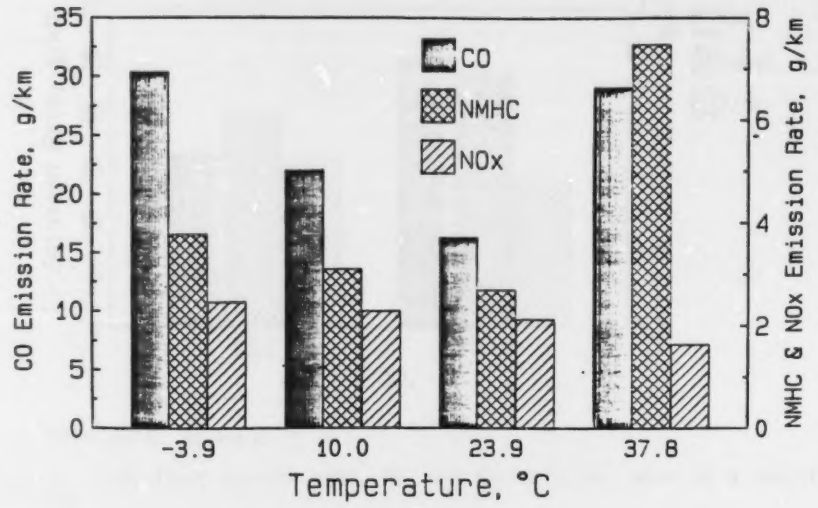


Note: 24°C, 152 m altitude

Fig. 7. 1985 fleet average NMHC, CO, and NOx emission rates as a function of speed.

speeds. The NMHC/NOx ratio varied from 2.2 at 8 km/h (5 mi/h) to 0.7 at 88 km/h (55 mi/h).

Fig. 8 illustrates how MOBILE 4 projects the sensitivity of emission rates to ambient temperature. The 1985 fleet average NMHC and CO emission rates were lowest at 24°C (75°F), whereas NOx emission rates continuously decreased as the temperature increased. The NMHC/NOx ratio is minimum at 24°C (75°F) and maximum at 38°C (100°F), 1.3 and 4.6, respectively. The elevation of the NMHC/NOx ratio at 38°C (100°F) occurs primarily because of increased evaporative HC emissions predicted by MOBILE 4. MOBILE 4 uses algorithms to adjust both tailpipe and evaporative emission rates for changes in ambient temperature. Tailpipe emission rates are adjusted over the entire



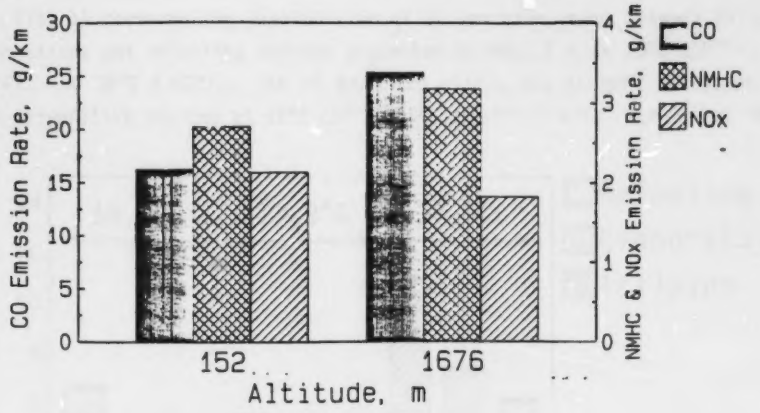
Note: 32 km/h, 152 m altitude

Fig. 8. 1985 fleet average NMHC, CO, and NOx emission rates as a function of ambient temperature.

summer-winter temperature range, and evaporative emission rates only in the summer range. Available data is inadequate to define evaporative emissions sensitivity to temperatures below 20°C (68°F). Likewise, available data is currently inadequate to define refueling emissions sensitivity to ambient temperature.

Fig. 9 illustrates how MOBILE 4 projects the influence of altitude on the characteristics of motor vehicle emissions. NMHC and CO emission rates are increased when vehicle altitude is increased, and NOx emission rates decreased. CO emission rates are most sensitive with 1985 fleet average values increasing 55% as the vehicle operating altitude is increased from 152 m (500 ft) to 1676 m (5500 ft).

The chemistry of tropospheric ozone formation depends critically on NMHC/NOx ratio (ref. 13). When this ratio is low the ozone forming potential is dependent on HC reaction rates, varying with the structure of the HC compound; but when this ratio is high the relatively low concentrations of NOx are limiting and the structure of the HC becomes less important. With motor vehicle emissions, this ratio varies with speed and temperature as indicated in Fig. 10. Over the scenarios examined, the highest ratio, 5.31, is predicted for 8 km/h (5 mi/h), 38°C (100°F) operating conditions, and the



Note: 24°C, 32 km/h

Fig. 9. 1985 fleet average NMHC, CO, and NOx emission rates as a function altitude.

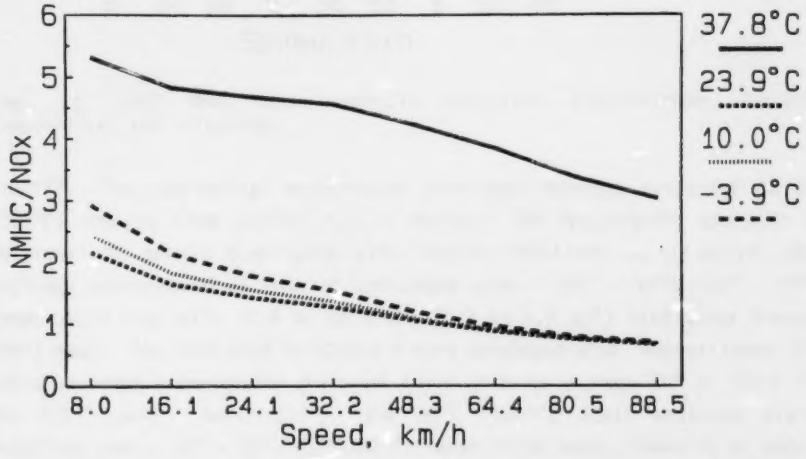


Fig. 10. 1985 fleet average NMHC/NOx ratios as a function of ambient temperature and average speed.

lowest ratio, 0.67, is predicted for 88 km/h (55 mi/h), 24°C (75°F) operating conditions. There is little sensitivity of this ratio to ambient temperature from -3.9°C (25°F) to 24°C (75°F) at 88 km/h (55 mi/h). The high values at 38°C (100°F) result primarily from large increases in evaporative hydrocarbon emission rates predicted by MOBILE 4.

Fig. 11 compares the distribution of HC emission rates between tailpipe, evaporative, and refueling sources projected by MOBILE 4 at 10°C (50°F), 24°C (75°F), and 38°C (100°F). At 32 km/h (20 mi/h), 34% of NMHC emissions were from evaporative sources at 10°C (50°F), 39% at 24°C (75°F), and 73% at 38°C

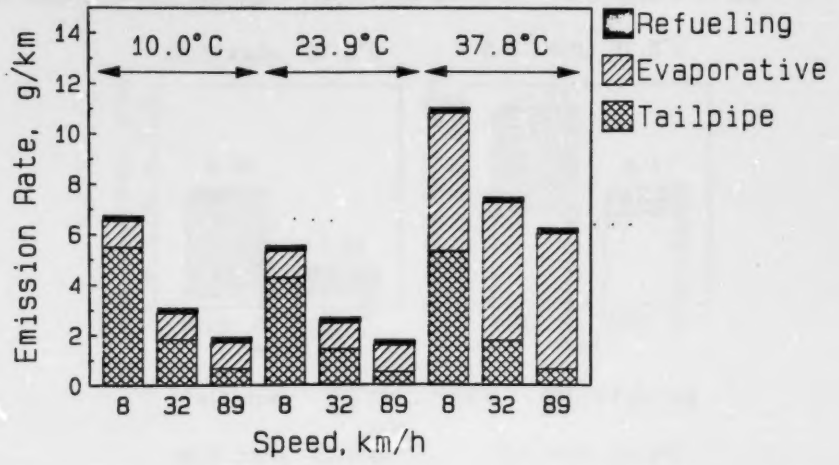


Fig. 11. 1985 NMHC motor vehicle emissions distribution: tailpipe, evaporative, and refueling.

(100°F). The substantial evaporative emissions increase projected at 38°C (100°F) results from control system design. The evaporative canister was developed to permit compliance with Federal Emissions Certification which includes measurement of diurnal emissions over a 16° - 29°C (60° - 84°F) temperature ramp with 60.0 to 63.4 kPa (8.7 to 9.2 psi) Reid Vapor Pressure (RVP) fuel. The data used in MOBILE 4 were developed with surveillance fleet vehicles using commercially marketed fuels with an average RVP of about 79.3 kPa (11.5 psi). Additionally, the 38°C (100°F) tests measured diurnal emissions over a 29° - 42°C (84°-108°F) temperature ramp. There is no data to support adjustment of evaporative or refueling emissions for vehicle average speed. However, since the evaporative control canister is regenerated while the engine is operated, and typically these devices do not purge while the engine is idling, evaporative emissions could be elevated when vehicles are operated at low average speed characterized by greater engine idle operation. With MOBILE 3 at 32 km/h (20 mi/h), 39% of HC emissions were projected from evaporative sources at 24°C (75°F), and 34% at 38°C (100°F). Fig. 12 compares MOBILE 3 and MOBILE 4 NMHC emission rates.

Tailpipe, evaporative, and refueling HC emissions are emitted at

different times and locations during normal vehicle operation. When considering the impact of motor vehicle emissions on atmospheric ozone, however, the aggregate of these sources must be considered. The composition of the VOC emissions from tailpipe, evaporative, and refueling sources are different and the aggregate composition will, therefore, depend on the rela-

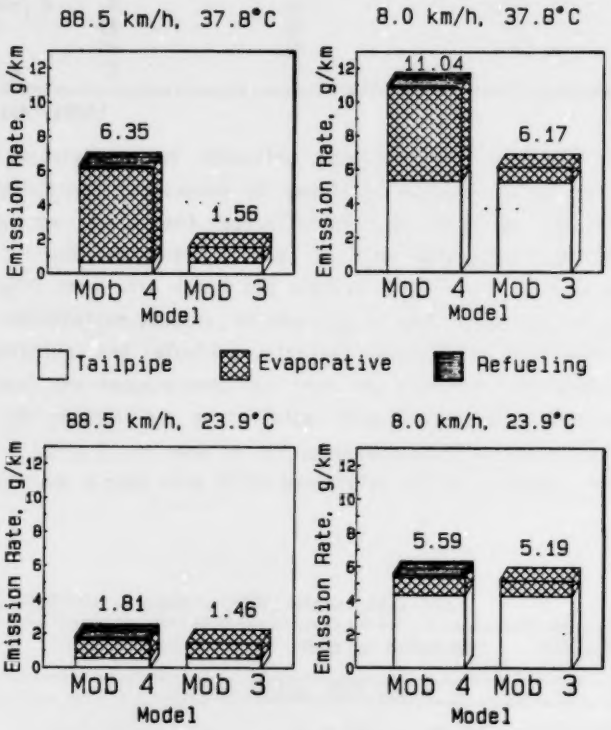


Fig. 12. Mobile 3 versus Mobile 4 NMHC emission rates.

tive amounts of each. Considering vehicle categorical emission strengths and VKT (see Table 2), light-duty gasoline vehicles, responsible for about 80% of total VOC, dominate the aggregate composition. The compositions of light-duty gasoline vehicle tailpipe, evaporative, and refueling VOC emissions have been examined and Table 3 provides data reflective of each source (ref. 14). The values presented are considered reasonable for currently marketed U.S. gasolines. The Motor Vehicle Manufacturers Association periodically publishes surveys of national gasoline characteristics as indicated in Table 4 (ref. 15). Emissions composition would be expected to vary as the range of gasoline composition.

TABLE 3.
Tailpipe, evaporative, and refueling hydrocarbon emissions composition

Hydrocarbon Classification	Tailpipe	Evaporative	Refueling
paraffinic, %	55	72	85
n-butane, %	5	23	32
isopentane, %	4	15	19
olefinic, %	18	10	11
aromatic, %	25	18	4
acetylenic, %	2	-	-

Source: Black (1988)

Both evaporative and refueling emissions are dominated by the more volatile paraffinic components of gasoline, e.g. n-butane and isopentane. Considering the fractional contributions of tailpipe, evaporative, and refueling sources indicated in Fig. 11, the composition of the aggregate emissions will vary with speed and ambient temperature as indicated in Fig. 13. This presentation assumes, at the risk of over simplification, that tailpipe, evaporative, and refueling emissions compositions are not highly sensitive to speed and temperature, but that the relative contributions of each are, i.e. the compositions of tailpipe, evaporative, and refueling emissions indicated in Table 3 are used in all speed-temperature scenarios. The aggregate compositions ranged from 70.8% paraffinic (21.7% n-butane, 14.1% isopen-

TABLE 4
Gasoline composition - summer, 1986, winter 1986/1987

	Regular Leaded	Regular Unleaded	Premium Unleaded
SUMMER FUELS, JULY 1987			
RVP, kPa[1]	71.0 (60.0 - 80.7)	71.7 (57.9 - 80.0)	73.8 (57.2 - 80.0)
Octane, $\frac{R+M}{2}$	88.6 (86.2 - 89.8)	86.9 (85.1 - 88.4)	91.8 (89.6 - 92.5)
Paraffinic, %	62.0 (55.6 - 68.3)	59.8 (54.0 - 65.5)	58.3 (50.4 - 67.7)
Olefinic, %	10.8 (6.6 - 15.0)	10.6 (7.3 - 16.1)	7.1 (3.7 - 13.5)
Aromatic, %	27.3 (23.9 - 31.3)	29.5 (25.7 - 33.3)	34.6 (26.4 - 40.6)
WINTER FUELS, JANUARY 1987			
RVP, kPa	93.1 (70.3 - 110.3)	93.1 (72.4 - 114.5)	92.4 (75.8 - 106.9)
Octane, $\frac{R+M}{2}$	88.6 (85.1 - 91.5)	87.3 (84.0 - 90.8)	91.8 (85.9 - 94.7)
Paraffinic, %	NA	62.1 (43.3 - 78.0)	59.9 (46.3 - 80.8)
Olefinic, %	NA	5.9 (2.2 - 16.6)	5.1 (1.0 - 14.0)
Aromatic, %	NA	32.0 (17.1 - 47.5)	35.0 (15.0 - 45.6)

[1] RVP - Reid Vapor Pressure: average (range)
Source: MVMA (1987)

tane), 10.8% olefinic, 18.2% aromatic, and 0.2% acetylenic hydrocarbon at 38°C (100°F), 88 km/h (55 mi/h) to 59.5% paraffinic (9.6% n-butane, 6.8% isopentane), 16.2% olefinic, 22.9% aromatic, and 1.4% acetylenic hydrocarbon at 24°C (75°F), 8 km/h (5 mi/h). The contribution of n-butane and isopentane ranged from 35.8% at 38°C (100°F), 88 km/h (55 mi/h) to 16.4% at 24°C (75°F), 8 km/h (5 mi/h). At lower temperatures, the aggregate would be more dominated by tailpipe emissions composition.

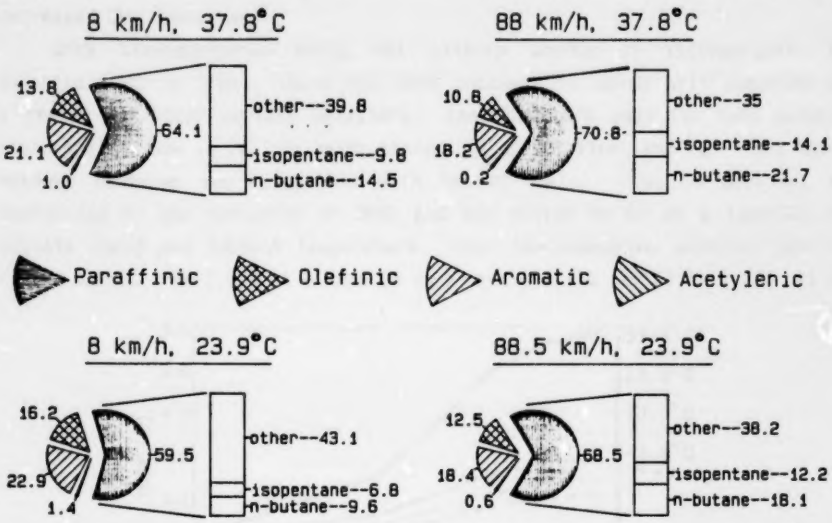


Fig. 13. Aggregate hydrocarbon emissions composition as a function of ambient temperature and average speed.

Methane, not being a component of gasoline, is emitted exclusively from the tailpipe during engine operation. Over the temperature and speed array of this presentation, the CH_4 fraction of tailpipe emissions is not highly variable for any selected calendar year. For example, during 1985 CH_4 constituted 5.69 ± 0.23% of tailpipe THC emissions over the entire temperature-speed array. Because of the varied contribution of tailpipe emissions to the total aggregate, the CH_4 fraction of the aggregate varied from 0.58% at 88 km/h (55 mi/h), 38°C (100°F) to 4.92% at 8 km/h (5 mi/h), -3.9°C (25°F). CH_4 is not reduced as effectively by catalyst control systems as other HC's, and therefore, typically constitutes a larger fraction of exhaust from well controlled cars. Thus, the CH_4 fraction of tailpipe and aggregate emissions increases with calendar year. During

1975 CH_4 constituted 5.00% of tailpipe and 2.95% of aggregate HC emissions at 32 km/h (20 mi/h), 24°C (75°F); and during the year 2000, it is projected to constitute 8.99% of tailpipe and 4.57% of aggregate HC emission at 32 km/h (20 mi/h), 24°C (75°F). The CH_4 fraction of tailpipe HC emissions from well controlled cars has also been reported to be sensitive to speed and ambient temperature (refs. 16, 17). The CH_4 fraction decreases with decreased average speed, and with decreased ambient temperature. With well controlled cars, operating conditions causing increased tailpipe THC emissions will result in decreased CH_4 fractions.

With transportation being the primary source of anthropogenic CO emission (70% in 1985), there has been interest in using this compound as a tracer for motor vehicle emissions. Lead has been used for this purpose historically, but is rapidly being phased out of gasoline, and fewer and fewer roadway vehicles are compatible with leaded fuels. Fig. 14 provides an indication of the variation of NMHC and NOx ratios to CO as a function of vehicle speed and ambient temperature. Over the scenarios examined NMHC/CO ratios varied from 0.098 at 8 km/h (5 mi/h) and -3.9°C (25°F) to 0.594 at 88

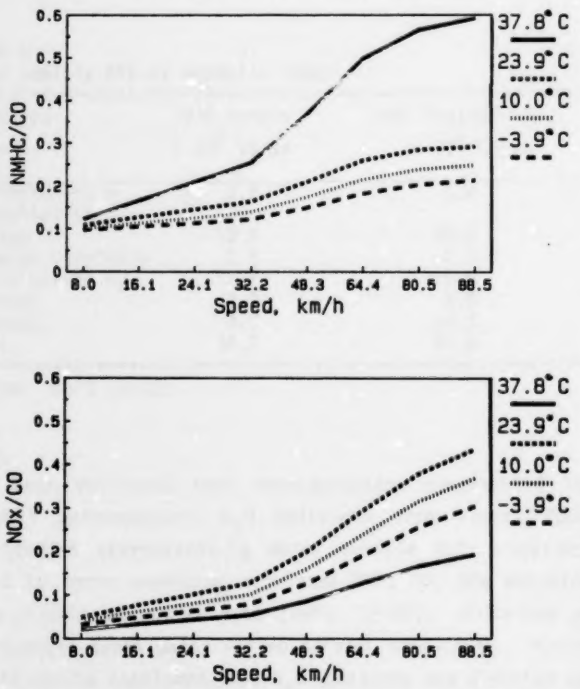


Fig. 14. 1985 fleet average NMHC/CO and NOx/CO ratios as a function of ambient temperature and average speed.

km/h (55 mi/h) and 38°C (100°F); and NOx/CO ratios varied from 0.023 at 8 km/h (5 mi/h) and 38°C (100°F) to 0.438 at 88 km/h (55 mi/h) and 24°C (75°F).

Chlorofluorocarbon emissions

The fully halogenated chlorofluorocarbons (CFC) most widely used in the U.S. are CFC-11 (CCl_3F), CFC-12 (CCl_2F_2), and CFC-113 ($C_2Cl_3F_3$). Motor vehicles were the single largest nonaerosol source of CFC in 1976, and are projected to be second to solvent emissions in 1990 (ref. 8). During 1976 29% of a combined 119 thousand metric tons emitted in the U.S. was from motor vehicle air conditioning systems; an additional 3.8% was from the production of flexible foam for motor vehicle applications. Emissions are projected to increase to 250 thousand metric tons during 1990 with 22% of this total from motor vehicle air conditioning systems. CFC-12 is exclusively used in motor vehicle air conditioning; CFC-11 is used in the production of flexible foam. Table 5 provides a summary of motor vehicle CFC-12 emission sources (ref. 18). Repair servicing and leakage were the most significant sources in 1976. Vehicle disposal is projected to become an additional significant source in 1990 projections.

TABLE 5
Motor vehicle CFC-12 emission rates

Emission Source	1976 Totals, x 10^6 kg/yr	1990 Projections, x 10^6 kg/yr
Manufacturing & installation	3.9	2.0
Leakage	10.3	16.5
Recharge servicing	1.5	2.1
Repair servicing	12.7	19.6
Accident	2.4	3.8
Disposal	3.9	11.3
Total	34.7	55.3

Source: Burt (1979)

N_2O emissions

It has been estimated that transportation contributed less than 10% of 1985 global anthropogenic N_2O emissions from fossil fuel combustion (ref. 7). Studies characterizing motor vehicle N_2O emissions are sparse when compared to those examining regulated THC, CO, NOx emissions. Table 6 summarizes available emissions data (refs. 19-25). Emissions were examined from varied vehicle types under varied driving conditions. Although the data is limited, it can be concluded that N_2O emissions are elevated about an order of magnitude from gasoline motor vehicles using catalysts compared to

noncatalyst configurations. Other observations in the available data include an increased N_2O emission rate as the vehicle average speed is decreased, if EGR (exhaust gas recirculation for NO_x control) is disabled, and as vehicle inertia weight is increased, i.e. truck emission rates are greater than passenger car emission rates. Using the vehicle categorical average emission factors of Table 6 and the 1985 fleet VKT distribution, a fleet average N_2O emission factor of about 30 mg/km can be estimated. This assumes that light-duty gasoline VKT is distributed 80-20 catalyst-noncatalyst. Coupled with total fleet kilometers traveled during 1985, highway motor vehicles emitted about 80 thousand metric tons of N_2O.

TABLE 6
Motor vehicle N_2O emission rates

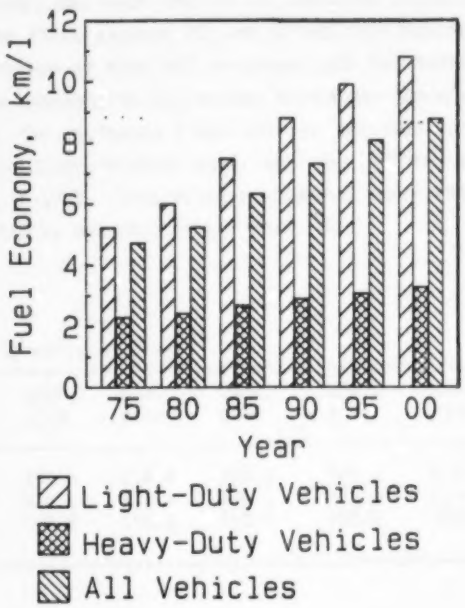
Vehicle Category	Estimated Emission factor, mg/km	Range, mg/km	References
Light-duty gasoline (no catalyst)	3.7	1.9 - 9.9	19,20
Light-duty gasoline (catalyst)	37.9	1.9 - 145.4	19,20,21,22,23
light-duty diesel	NA	6.8 - 29.8	19
heavy-duty gasoline	45.4	29.8 - 60.3	24
heavy-duty diesel	29.2	19.3 - 46.6	24, 25

The data base was examined for relationships between N_2O emissions and NOx emissions. The observed N_2O/NOx ratios were 0.0437 ± 0.0414 (n=38) for catalyst equipped gasoline vehicles, 0.0018 ± 0.0002 (n=5) for diesel trucks, and 0.0055 ± 0.0028 (n=2) for gasoline trucks (no catalysts). The "catalyst equipped" data base included more test vehicles and a larger array of driving conditions, i.e. speed, ambient temperature, malfunction, and mileage accumulation, than the truck data base. The range of observed N_2O/NOx ratios was therefore greater than observed with the trucks. There is no certainty that the trucks will exhibit the same sensitivity to these variables. As the world's vehicle fleet shifts to catalyst technology for abatement of urban ozone, CO and NOx, the contribution of transportation sources to global N_2O burden will grow.

Carbon dioxide emissions

As previously mentioned, highway motor vehicles were estimated to be responsible for about 24% of CO_2 emissions from 1985 fossil fuel combustion in the U.S. (ref. 10). The emission rate of CO_2 from motor vehicles is a

function of fuel economy and fuel characteristics, i.e. liters of fuel consumed per kilometer driven, and grams of carbon per liter of fuel. Significant fuel economy improvements have been realized in the U.S. in recent years as a consequence of the combined effects of market demand and federal regulations. Fig. 15 is illustrative of fuel economy improvements in both light-duty and heavy-duty vehicle categories since 1975 with projections to the year 2000 (ref. 3).



Source: Mobile 3 Fuel Consumption Model
EPA-AA-TEP-EF-85-2, Feb. 1985

Fig. 15. Motor vehicle fuel economy, 1975 to 2000.

The two major transportation fuel categories, gasoline and diesel, typically have 619 to 654 and 706 to 741 grams of carbon per liter, respec-respectively. Most of this carbon is converted to CO_2 with lesser amounts to CO and HC during combustion in motor vehicle engines. CO_2 emission rates can be calculated from fuel economy data according to the carbon balance relationship:

$$CO_2, g/km = ((K_1 x FD, g/\ell) / FE, km/\ell - K_1 x HC, g/km - K_2 x CO, g/km) / K_3 \quad (3)$$

where: K_1 = carbon weight fraction of fuel = .866; K_2 = carbon weight fraction of CO = .429; K_3 = carbon weight fraction of CO_2 = .273; FD = fuel density; and FE = fuel economy.

Combining MOBILE 4 THC and CO emissions data with fuel economy data, CO_2 emission rates can be calculated as presented in Table 7. The trend, with improved fuel economy, has been reduced CO_2 emission rates. During the 1975 to 1985 decade, the fleet average CO_2 emissions rate was reduced about 22%. During that same period of time VKT increased 30% (as indicated in Fig. 1), thus increasing the atmospheric CO_2 burden from motor vehicles on roadways in the U.S. Combining the estimated fleet average emission rate with estimated nationwide miles traveled, highway motor vehicles emitted about 0.9 billion metric tons of CO_2 in 1985. This value is somewhat lower than the 1.1 billion metric tons estimated by DeLuchi, et al. (ref. 10).

TABLE 7
Motor vehicle CO_2 emission rates

Year	LDGV, g/km	LDGT, g/km	HDGV, g/km	LDDV, g/km	LDDT, g/km	HDDV, g/km	ALL g/km
1975	352.4	395.8	614.4	283.3	204.2	1293.7	426.2
1985	270.0	330.8	532.5	236.1	271.0	1076.1	334.1
1995	208.8	284.8	555.3	199.5	252.0	908.1	268.2

SUMMARY AND CONCLUSIONS

Stratospheric and tropospheric ozone concentrations are controlled by complex chemical and physical processes which are influenced by both anthropogenic and biogenic emissions at the earths surface. One of the most rapidly growing human activities important to atmospheric ozone is the use of highway motor vehicles. Miles driven by American motorist have increased 58% since 1970. There are many compounds emitted from highway motor vehicles that participate in atmospheric processes important to ozone formation and/or destruction including VOC, CO, NOx, CO_2, N_2O, and CFC-12. It has been estimated that during 1985 7.2 million metric tons of VOC were emitted from U.S. transportation sources (about 34% of the anthropogenic total), 47.5 million metric tons of CO (about 70% of the anthropogenic total), and 8.9

million metric tons of NOx (about 45% of the anthropogenic total). Recent improvements to MOBILE 3, the model used to calculate fleet average emission factors, will increase the estimated transportation contribution to VOC emissions to nearly 50% of the anthropogenic total. Summer month evaporative hydrocarbon emission estimates are significantly increased. U.S. motor vehicles were also estimated to have been responsible for about 48 thousand metric tons of CFC during 1985 (25% of U.S. anthropogenic total), and about 1 billion metric tons of CO_2 (14% of the anthropogenic total). Motor vehicles contribute less than 10% of anthropogenic N_2O emissions, but this contribution is expected to grow as the fraction of catalyst equipped vehicles increases.

Through the cooperative efforts of government and industry, substantial reduction of motor vehicle emissions has been achieved during the past decade. From 1975 to 1985, motor vehicle VOC emission rates were reduced about 57%, CO rates about 62%, NOx rates about 35%, and CO_2 rates about 22%. Much of this reduction was offset, however, by a concurrent 30% growth of VKT.

Motor vehicle emission rates are sensitive to driving conditions such as speed, temperature, and altitude. Generally, VOC and CO emission rates are increased by reduced average speed. NOx emission rates, not as sensitive to speed, are generally minimumized at about 32 to 48 km/h and increase slightly at lower and higher average speeds. VOC and CO emission rates are minimumized at about 24°C (75°F) and increase at lower and higher temperatures. NOx emission rates increase as ambient temperature is reduced. Typically, VOC and CO emissions rates increase as altitude is increased, and NOx emission rates decrease. These variations influence the NMHC/NOx ratio of the emissions, an important consideration to tropospheric ozone chemistry. This ratio varies from about 0.7 to 5.3 over the examined range of driving conditions.

VOC emissions from motor vehicles originate from tailpipe, evaporative, and refueling sources. The composition of each is different, with the more volatile C_4 and C_5 paraffins dominating evaporative emissions and, even more so, refueling emissions. The aggregate emissions composition will depend on the relative contribution of each, a function of vehicle speed and ambient temperature. At 24°C (75°F), 8 km/h (5 mi/h), 1985 motor vehicle VOC emissions were about 77% tailpipe, 19% evaporative, and 4% refueling; at 38°C (100°F), 88 km/h (55 mi/h) the emissions were about 10% tailpipe, 86% evaporative, and 4% refueling. The aggregate composition was 60% paraffinic, 16% olefinic, 23% aromatic, and 2% acetylenic at 24°C (75°F), 8 km/h (5 mi/h); and 71% paraffinic, 11% olefinic, 18% aromatic, and 1% acetylenic at 38°C (100°F), 88 km/h (55 mi/h).

CFC-12, the primary chlorofluorocarbon emitted from motor vehicles, results primarily from air conditioning repair service and leakage. Vehicle disposal is projected to become a significant source by the 1990's. U.S. motor vehicle CFC emissions are projected to be about 55 thousand metric tons per year by 1990. N_2O emissions from motor vehicles have not been widely studied, but available data suggests that emission rates vary over about an order-of-magnitude range from about 3.7 mg/km from noncatalyst gasoline passenger cars to about 37 mg/km from catalysts equipped gasoline passenger cars. Limited data suggests that heavy-duty truck emission rates are greater than passenger car emission rates (with similar engine emission control configurations). Fleet average CO_2 emission rates were reduced from about 426 g/km in 1975 to about 334 g/km in 1985.

Further emission rate reduction is possible through curtailment of vehicle tampering and misfueling, and improved maintenance. Additionally, the U.S. Congress is considering expanded regulation of tailpipe, evaporative, and refueling emissions, and fuel composition. Ultimately, as the potential of emission rate reduction becomes realized, only curtailment of VKT growth will be available to limit motor vehicle emissions.

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